

Tailoring Dielectric Properties of Polymer Matrix Composites for High-performance Flexible Sensors

Jun Li^{a,b,*}, Kit-Ming Ma^b, Li-Sha Zhang^{b,*}, and Xiao-Ming Tao^{b,*}

^a School of Environmental and Chemical Engineering, Wuyi University, Jiangmen 529020, China

^b Research Institute for Intelligent Wearable Systems, School of Fashion and Textiles, The Hong Kong Polytechnic University, Hong Kong 999077, China

Electronic Supplementary Information

Abstract Polymer matrix composites with high dielectric constants and low dielectric losses are in high demand for flexible electronics. However, simultaneously satisfying these requirements poses a significant scientific challenge owing to the intrinsic trade-off relationship. Herein, we utilized the *in situ* controllable reduction of graphene oxide (GO) within a poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene) (P(VDF-TrFE-CFE)) matrix to regulate the dielectric properties. The as-obtained composite exhibited a high relative dielectric constant of 1415 coupled with a low loss tangent of 0.380 at 100 Hz. Experimental and theoretical studies indicate that the increased degree of electron conjugation and conductivity of the reduced GO (RGO) are responsible for the high-*k*. The constrained reduction degree of GO, combined with its homogeneous dispersion in the polymer matrix, effectively suppresses long-range charge carrier migration, thereby minimizing dielectric loss. This novel strategy could be successfully applied to both organic and aqueous systems. Furthermore, a high-performance flexible capacitive proximity sensor was exemplified by the optimization of both the dielectric layer and electrode pattern, exhibiting excellent sensitivity and stability. The fundamental mechanisms elucidated in this study provide crucial design principles for developing dielectric PMCs with tailored properties, thereby opening new avenues for advanced flexible electronic applications.

Keywords Dielectric property regulation; High-*k*; Polymer matrix composites (PMCs); Capacitive proximity sensor

Citation: Li, J.; Ma, K. M.; Zhang, L. S.; Tao, X. M. Tailoring dielectric properties of polymer matrix composites for high-performance flexible sensors. *Chinese J. Polym. Sci.* 2026, 44, 116–126.

INTRODUCTION

Dielectric materials, which are key components of electronic devices, have drawn increasing technological and scientific interest in recent years.^[1–3] There is an urgent demand for the development of flexible dielectric materials with high dielectric constants (high-*k*) and low dielectric losses, particularly for applications in actuators,^[4] optoelectronics,^[5] communication technology,^[6] energy storage,^[7,8] energy harvesting,^[9,10] and sensors.^[11,12] Compared with traditional rigid ceramic or inorganic dielectric materials, their inherent flexibility makes polymer-based high-*k* systems good candidates for flexible and wearable electronics.^[5,13,14] Pristine polymers usually exhibit relatively low dielectric constants, although molecular modifications have been demonstrated to augment the properties several times.^[15–17] Polymer matrix composites (PMCs) are used to develop high-*k* systems by incorporating high-*k* materials^[18–20] or conductive fillers^[21,22] into the polymer matrix. Doping with

conductive fillers can increase the dielectric constant by several orders of magnitude near the percolation threshold, which is attributed to the enhanced interfacial polarization in the composites.^[22] Interfacial polarization is also known as the Maxwell-Wagner-Sillars (MWS) effect, which is associated with the accumulation of polarized charges at the conductive filler/insulative polymer matrix interface. This effect is more pronounced in the low-frequency range. Carbon-based conductive fillers demonstrate superior chemical and mechanical compatibility with polymer matrices compared with metals.^[23,24] Among these, flake-structured graphene is a promising nanofiller because of its anisotropic features.^[22] However, aggregation and stacking cannot be avoided between adjacent graphene nanosheets owing to the large surface free energy, which suppresses the dielectric constant and leads to significant dielectric loss.^[25] The elevated loss in dielectrics leads to high power consumption, overheating, and even device failure.^[21,26]

Numerous studies have been conducted to minimize the dielectric loss owing to dielectric relaxation and leakage current while maintaining a high-*k*. The most widely used methodology is to modify the surface properties of fillers. A core-shell structure is generally adopted, where an insulative shell (composed of metal oxides or polymers) can be coated onto the conductive or ceramic core through grafting, calci-

* Corresponding authors, E-mail: lijun@wyu.edu.cn (J.L.)

E-mail: lisa-lisa.zhang@polyu.edu.hk (L.S.Z.)

E-mail: xiao-ming.tao@polyu.edu.hk (X.M.T.)

Received June 4, 2025; Accepted September 30, 2025; Published online December 18, 2025

nation, deposition, and other surface engineering technologies.^[21,27–29] Zhou *et al.* recently proposed a domain-type polarization mechanism for high-*k* PMCs with core-shell fillers, wherein each domain comprised a cluster of conductive fillers instead of a single particle.^[21] A higher-than-normal concentration of core-shell fillers with optimized shell electrical property charge transport among clusters. The mechanical and chemical compatibility with the polymer matrix can also be improved by proper surface modification of fillers. The homogenous dispersion and ordered alignment of the fillers could also suppress the formation of conductive pathways and thus suppress dielectric loss. Sandwich or multilayer structures are also an effective method to obtain low-loss PMCs, especially for energy storage owing to the ohmic barrier effect, although the dielectric constant may not be high enough.^[30–32] PMCs with highly aligned aerogel frameworks prepared by unidirectional freeze casting^[33,34] or 3D networks^[35] represent another effective strategy for preparing high-dielectric-constant composites with low dielectric loss, although complicated preparation processes are typically involved. It can also be considered a multilayer structure based on conductive aerogel frameworks and insulating polymer barriers. In summary, despite extensive efforts have been made in this area, obtaining high-dielectric-constant composites with low dielectric losses remains a challenge.

In this work, a straightforward strategy for a high-dielectric-constant composite with low dielectric loss is proposed. A high-*k* PMC based on a partially reduced graphene oxides (PRGO)/poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene) (P(VDF-TrFE-CFE)) composite with tunable dielectric properties was successfully prepared through *in situ* thermal reduction of GO in a polymer matrix. A remarkable ϵ_r (relative dielectric constant) of 1415 and $\tan\delta$ (dissipation factor) of 0.380 at 100 Hz were achieved, outperforming most dielectric PMCs. Experimental and theoretical studies reveal that a high-*k* is caused by the enhanced conjugation degree after reduction, and the dielectric constant increases monotonically with increasing conductivity of GO. Concurrently, the uniform dispersion of graphene oxide (GO) within the polymer matrix, coupled with the inhibited long-range migration of charge carriers due to the limited reduction degree of GO, minimizes the leakage current and is responsible for the low dielectric loss. This regulation strategy has also been successfully applied to water-soluble polymer matrices, such as PVA, indicating its wide applicability. Finally, a high-performance planar structured capacitive proximity sensor with excellent sensitivity and stability was fabricated with an optimized electrode pattern using substantial numerical simulations. This study holds significant importance for advancing flexible dielectric materials and is poised to make substantial contributions to practical applications in wearable electronic devices.

EXPERIMENTAL

Procedures for the Sample Preparation

PRGO/P(VDF-TrFE-CFE) composite: a 4 wt% PRGO/P(VDF-TrFE-CFE) composite was used as an example. 40 mg of GO and 1.00 g of P(VDF-TrFE-CFE) powder was separately fully dissolved in 4 and 10 mL of *n*-butyl acetate (BA) solution and both mixtures

were stirred for at least 4 h. The two solutions were then mixed and stirred for approximately 6 h to obtain a homogeneous 4 wt% GO/P(VDF-TrFE-CFE) solution. Then, blade coating on Al foil or other substrates was conducted on a *k* control coater with profile bars. The film was dried at 60 °C under vacuum for 2 h to remove the solvents, followed by thermal reduction at 120 °C for different times to produce the PRGO/P(VDF-TrFE-CFE) films. The temperature was slightly lower than the melting point of P(VDF-TrFE-CFE) (about 127 °C) to retain the homogeneous dispersion of GO in the polymer matrix. For other doping ratios of GO composites, the only difference was the weight of GO; it should be noted that the GO must be dispersed first; otherwise, it could not be successfully dispersed with the increased solution viscosity.

RGO film: pure RGO samples were also prepared for comparison, and typically 100 μ L of GO solution (10 mg/mL in BA) was dropped on a clean glass substrate and then dried and thermally reduced under the same conditions as the GO/P(VDF-TrFE-CFE) samples.

Vitamin C (V_C)-RGO/P(VDF-TrFE-CFE) composite: for chemical reduced GO/P(VDF-TrFE-CFE) composite, the GO/P(VDF-TrFE-CFE) film was immersed into an ascorbic acid (V_C) solution (5 mol·L⁻¹) and the solution was kept at 90 °C for 0.5 h to reduce the GO. The V_C -RGO/P(VDF-TrFE-CFE) film was then rinsed with deionized (D.I.) water for three times to remove impurities and finally dried at 80 °C for 1 h.

P(VDF-TrFE-CFE) film: a 0.1 mg/mL BA solution of the polymer was used for the blade coating, which is the same as that of the composite. The film was dried at 60 °C under vacuum for 2 h to remove the solvents, followed by thermal annealing at 120 °C overnight to increase crystallinity.

PRGO/PVA composites: 20 mg of GO and 1.00 g of PVA were fully dissolved in 2 and 10 mL of D.I. water for at least 4 h, and the two solutions were mixed and stirred for 6 h. The film was prepared by solution casting and dried at 60 °C under vacuum for 2 h to remove the solvents, followed by thermal reduction at 160 °C for different durations.

Procedures for the Sensor Fabrication and Testing

The electrode was prepared through mask-assisted vacuum deposition on a PI substrate, and the high-*k* layer was screen printed directly on the flexible electrode. For the cycle stability measurement, the sensor was placed on Instron 5944 with an iron rod on the load cell as the sensing target. The load cell moved back and forth in the distance range from 5 mm to 100 mm, and the capacitance change was recorded using an impedance analyzer. All measurements were conducted under a constant conditions (21 °C and relative humidity of about 70%).

Finite Element Analysis

Three-dimensional (3D) models were designed and constructed using the AC/DC module in COMSOL Multiphysics® 6.1. The material properties and boundary conditions of the models were based on the experimental results. Two initial assumptions were made: (1) the dielectric material ideally adhered to the electrodes; (2) except for the sensor, material under test (MUT), and air, nothing was in space. The mesh was controlled by the users. An unstructured quadrilateral mesh, known as a free quad mesh, was generated on the surface of the sensors and MUT with the limitation of a maximum element size of 0.1 cm. The free quad-mesh was swept through the sensors. A free tetrahe-

dral mesh was used for air. This study was conducted using a stationary source sweep.

RESULTS AND DISCUSSION

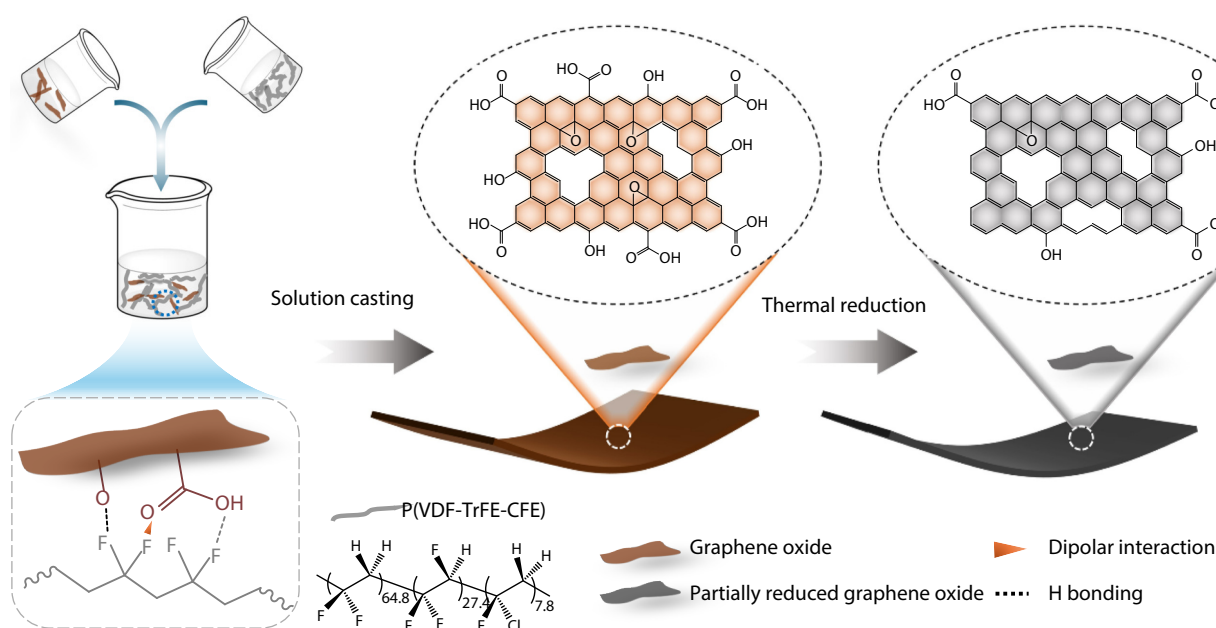
High-*k* PMC with Tunable Dielectric Properties

A PRGO/P(VDF-TrFE-CFE) composite with tunable dielectric properties was prepared through controlled *in situ* thermal reduction, as illustrated in Scheme 1. With a PRGO doping ratio of 4 wt%, the composite could provide a high dielectric constant of 1415 with a relatively low dielectric loss ($\tan\delta$) of 0.380 at 100 Hz, exceeding the performance of most dielectric PMCs. The abundant hydrogen bonding and dipolar interactions between the residue groups of GO and polymer chains enable uniform nanofiller dispersion,^[36] which is beneficial for obtaining a high-*k* and low loss. The reduction in the time-dependent dielectric properties of the composite is shown in Figs. 1(a)–1(c) and Table S1 (in the electronic supplementary information, ESI). It was observed that both the dielectric constant and loss increased monotonically with reduction time until equilibrium was reached at a relatively low reduction temperature of 120 °C. The set temperature was slightly lower than the melting point of the polymer, which helped maintain a good dispersion of GO within the polymer matrix. Day 0 corresponds to the as-prepared samples, which were dried at 80 °C under vacuum for 2 h, and Day 1–14 means the dried samples were thermally reduced at 120 °C under vacuum for the corresponding time. Both the dielectric constant and loss remained almost unchanged on Day 13 and 14, indicating that GO was fully reduced after two weeks under the abovementioned conditions. The interfacial polarization between the nanofillers and polymer is dominant in the low-frequency regime (below 10^3 Hz), which arises from the improved conductivity and interfacial polarization associated with the reduced GO. The intensity of interfacial polarization decreased dramatically with increasing frequency. Meanwhile, the dielectric loss also increases with the reduction degree, particu-

larly in the low-frequency region, and tends to rise again in the high-frequency region ($>10^3$ Hz), which is attributed to charge-carrier hopping and relaxation processes. A gradual downward trend was observed in the loss of the pure polymer in the measured frequency range, which is typical for polar polymers. However, for the composite samples, an increasing trend was observed, and the peak shifted to the high-frequency range with reduction time. This can be ascribed to the dominant ion polarization/conductance in the high-frequency range of the conductive fillers in response to the electric field.

The obtained high-*k* PMC also exhibited excellent stability because the dielectric properties of the composite remained unchanged under ambient conditions for more than 30 days, as demonstrated in Fig. 1(d). The breakdown strength is also a key dielectric parameter that determines the safety of operation, especially when applied as a dielectric layer in capacitors. The breakdown strength (E_b) and Weibull distribution of the 4 wt% PRGO/P(VDF-TrFE-CFE) composite were characterized according to literature.^[37–39] The E_b was measured to be (5.58 ± 0.30) V/ μm , and the Weibull distribution is shown in Fig. S2(d) (in ESI). The steep slope ($\beta=18.53$) indicates a narrow distribution of E_b , suggesting good consistency in the sample preparation.

Owing to the superior dispersibility of GO in both aqueous and organic solvents, manipulation of the dielectric properties in aqueous systems can be feasibly realized as well. This dielectric property control strategy is applicable to aqueous PRGO/PVA systems, as shown in Figs. 1(e)–1(g). The reduction could reach equilibrium within 1 h at an elevated reduction temperature of 160 °C. The 4 wt% PRGO/PVA composite exhibits a relative dielectric constant of 227 and loss of 1.01 at 100 Hz; while for pristine PVA, they are 3.6 and 0.07, respectively. An increased reduction temperature is expected to shorten the time required to reach thermodynamic and chemical equilibria, as demonstrated in previous studies.^[40–42]



Scheme 1 Schematic illustration of the PRGO/P(VDF-TrFE-CFE) PMC preparation.

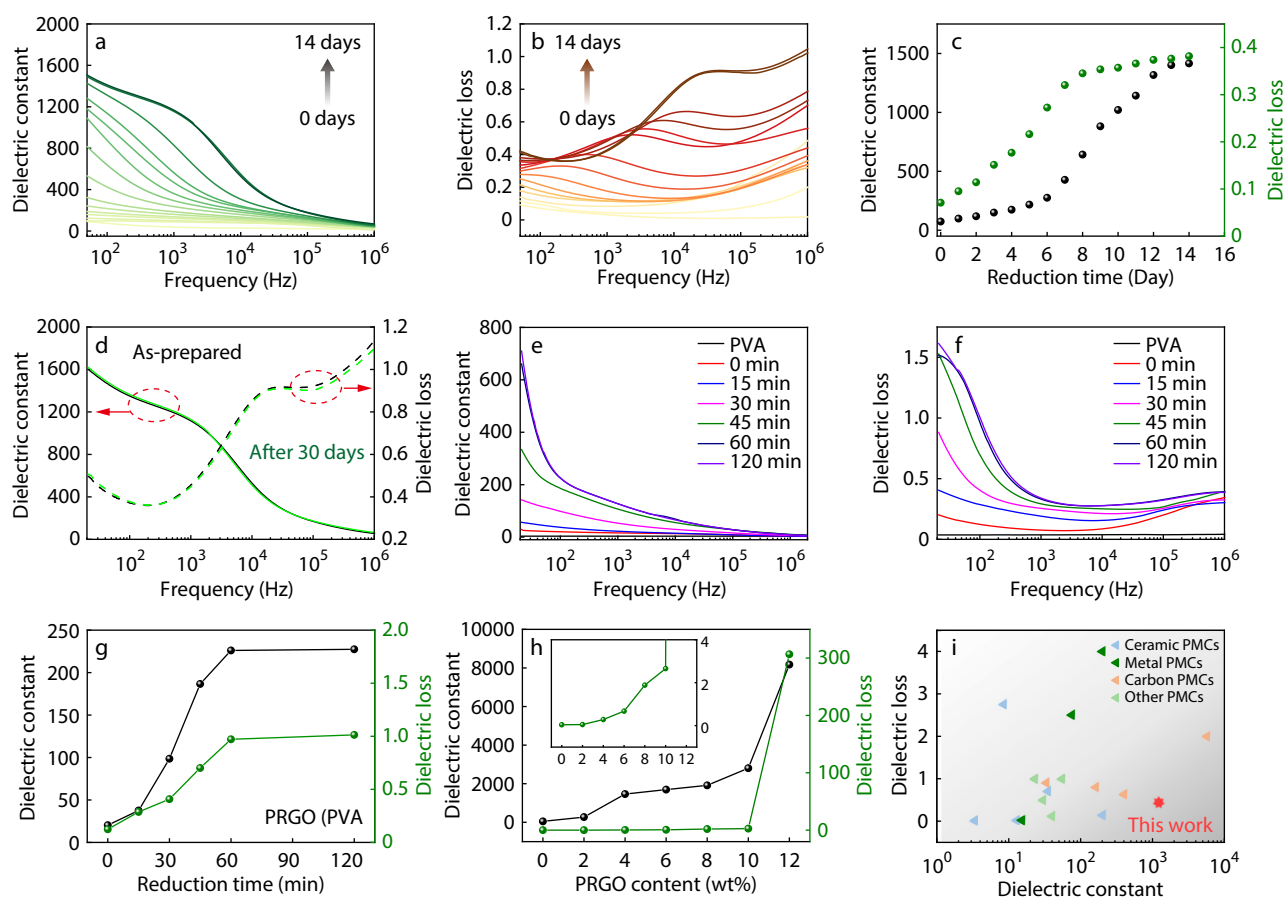


Fig. 1 Dielectric properties of the PMCs. (a) Dielectric constant and (b) dielectric loss with different reduction time (at 120 °C) of 4 wt% GO doping in P(VDF-TrFE-CFE) as a function of frequency; (c) Dielectric constant and dielectric loss in GO/P(VDF-TrFE-CFE) composites as a function of reduction time at 100 Hz; (d) Stability of the high- k composites after 30 days; (e) Dielectric constant and (f) dielectric loss with different reduction time (at 180 °C) of 4 wt% GO doping in PVA as a function of frequency; (g) Dielectric constant and dielectric loss in GO/PVA composites as a function of reduction time at 100 Hz; (h) Percolation threshold of GO/P(VDF-TrFE-CFE) composites; (i) Benchmarks of high- k PMCs with conductive or ceramic fillers.

The dielectric properties of PMCs with different PRGO doping ratios were investigated following the same preparation procedures. The results are presented in Fig. S1 and Table S2 (in ESI). The relative dielectric constant reaches about 8100 at 100 Hz with a GO doping ratio of 12 wt% though $\tan\delta$ is higher than 300. A typical percolative transition was observed at about 12 wt%.^[5,43] The direct current (DC) sheet resistance of the 12 wt% PRGO/P(VDF-TrFE-CFE) was measured to be $(51.93 \pm 2.84) \text{ k}\Omega\text{-cm}/\square$, indicating that conductive pathways may form in the matrix. The alternating current (AC) conductivities of the composites were also measured, and the data are summarized in Fig. S2 (in ESI). Figs. S2(a) and S2(b) (in ESI) show the real part (σ_{AC}') and imaginary part (σ_{AC}'') of the AC conductivity of the PRGO/P(VDF-TrFE-CFE) composite. The AC conductivity exhibited only a slight variation in the low-frequency region ($<1 \text{ kHz}$), indicating limited carrier mobility in slowly varying fields. With increasing frequency, a gradual increase in conductivity was observed, which can be attributed to the more effective hopping or tunneling of charge carriers within the heterogeneous microstructure. Such frequency-dependent behavior is consistent with typical conductive-filler/polymer-based nanocomposites.^[44,45] Interestingly, it

could be found the 2 wt% and 4 wt% composites exhibited higher AC conductivity than those of the 6 wt% and 8 wt% samples. This can be explained by the more uniform dispersion of RGO sheets at low concentrations, which maximizes interfacial polarization and enhances charge hopping or tunneling under alternating fields. The imaginary part (σ_{AC}'') of the 2 wt% and 4 wt% composites indicates a typical transition from capacitive to inductive response. While a positive σ_{AC}'' generally corresponds to capacitive polarization, a negative σ_{AC}'' implies the dominance of an inductive-like impedance or delayed interfacial relaxation at high frequencies. The decrease in σ_{AC}'' with frequency arises because interfacial polarization cannot follow the rapidly alternating field, leading to suppressed capacitive storage and an enhanced phase lag of the charge carriers.

The dielectric constants and dielectric losses of the samples at 100 Hz are displayed in Fig. 1(h), which could provide a more intuitive concept for the percolation transition. The dielectric constant and loss did not exhibit an obvious change from 4 wt% to 10 wt%. However, at a doping ratio of 4 wt%, which is well below the percolation threshold, the dielectric loss was significantly lower than that at 10 wt% (see Fig. 1h

and Table S2 in ESI). The low doping ratio also facilitates uniform dispersion and minimizes the leakage current within the composite, thus reducing dielectric loss. A benchmark for high- k composites with conductive or ceramic fillers is shown in Fig. 1(i) (the bottom-right area is preferred), and more detailed information is provided in Table S3 (in ESI). It is clear that the *in situ* controlled thermal reduction of GO in the polymer matrix provides a feasible strategy to obtain high- k PMCs with low loss.

Mechanism Study

The 14-day conductance (G) change of PRGO under the same conditions was also recorded (see Fig. S3 in ESI). The normalized conductivity and ϵ_r are shown in Fig. 2(a). Interestingly, the growth trends of ϵ_r and G demonstrate a notable degree of similarity. This result indicates that the dielectric properties can be monitored and estimated by conductance in a nondestructive manner. The conductance is closely related to the extent of PRGO reduction, which varies with the reduction time. The main reason for the increase in the dielectric constant and loss can be ascribed to the enhanced conductivity of the PRGO.^[43,46] This methodology enables precise control of the dielectric constant

in the material processing procedure, providing the possibility for on-demand dielectric material design for specific requirements.

Although we already know how to harness the dielectric properties of a composite, it is crucial to understand the underlying mechanism. The FTIR spectra of pure P(VDF-TrFE-CFE), GO powder, 4 wt% GO/P(VDF-TrFE-CFE), and 4 wt% PRGO/P(VDF-TrFE-CFE) are shown in Fig. 2(b). Symmetric stretching of the all-trans (β phase) $-\text{CF}_2$ and $-\text{CH}_2$ vibrations can be observed at 846 and 1398 cm^{-1} , respectively, and the band at 770 cm^{-1} arises from the γ phase of P(VDF-TrFE-CFE).^[47] In the 4 wt% GO/P(VDF-TrFE-CFE) composites, two bands at 2917 and 2849 cm^{-1} were observed, which are typical bands for C—H vibrations from the $-\text{CHO}$ groups of GO. The bands at 1625 and 1723 cm^{-1} in the GO powder were assigned to the C=O from GO,^[48] but shifted to 1578 and 1541 cm^{-1} in the composites. The large blue shift can be ascribed to the hydrogen bonding and dipolar interactions between the filler and polymer,^[36] which redistribute the electrons of C=O double bonds. A similar shift was reported for the H-bonded PRGO/PEG/TPU composites.^[49] These two bands in

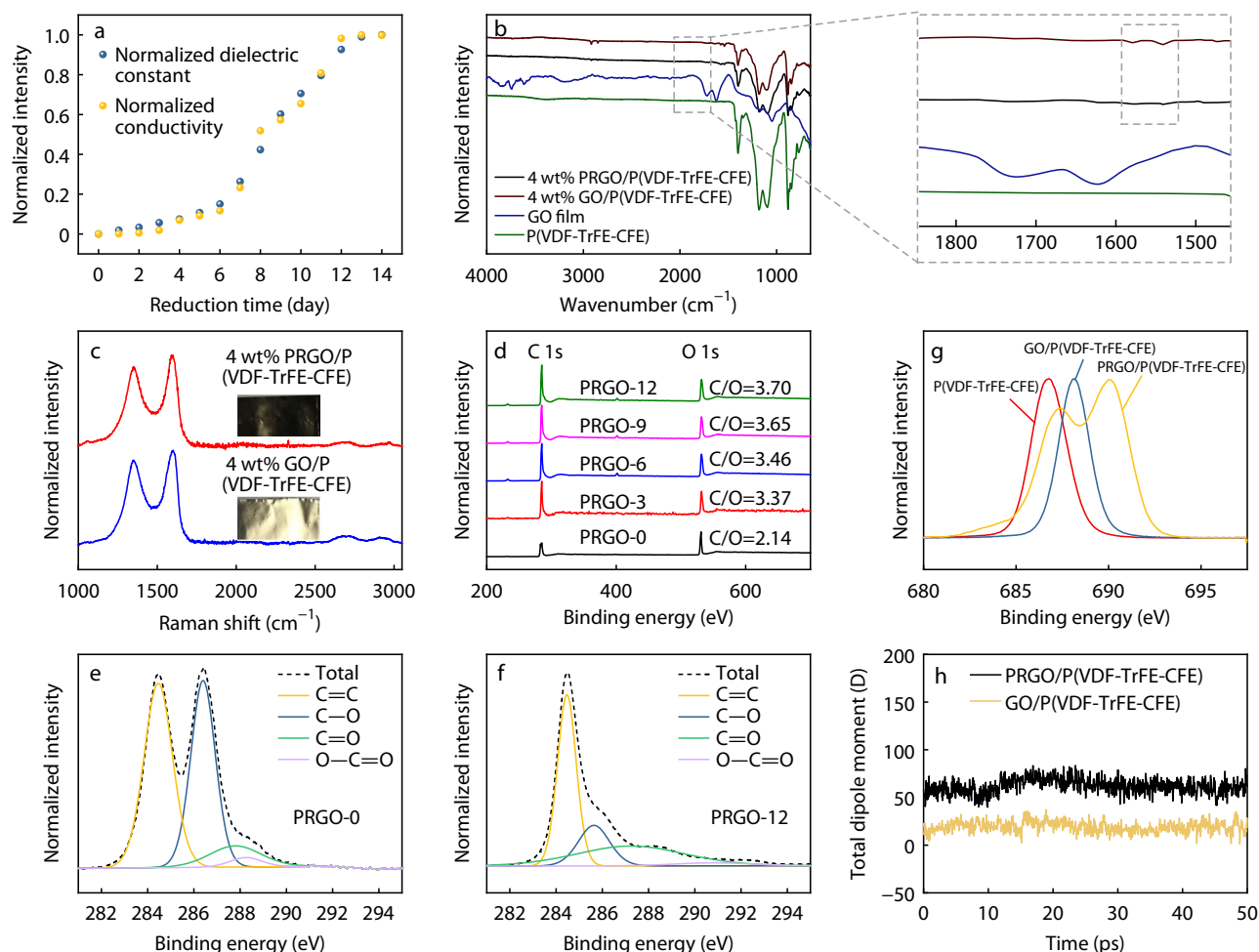


Fig. 2 Characterization and mechanism study. (a) Normalized conductivity of the GO films and normalized dielectric constant with different reduction time; (b) FTIR and (c) Raman spectra of the composites; (d) XPS spectra of PRGO powders with different reduction time; fitting results of C 1s spectra of (e) PRGO-0 and (f) PRGO-12; (g) F 1s XPS spectra of P(VDF-TrFE-CFE), GO/P(VDF-TrFE-CFE) and PRGO/P(VDF-TrFE-CFE) films; (h) MD simulation results of the dipole moments in GO/P(VDF-TrFE-CFE) and PRGO/P(VDF-TrFE-CFE) composites.

the reduced composites at the same wavenumbers were observed to have lower intensities, indicating that the oxygen-containing functional groups in GO were partially removed after thermal treatment, and the interactions between PRGO and the polymer still existed. The shift of the dielectric loss peak in frequency may also be related to the change in the filler-polymer interactions with an increased degree of reduction. The uniform dispersion of GO in the polymer matrix is reasonable for achieving low-loss characteristics, where the formation of the potential conductive pathways will be significantly suppressed, thus reducing the leakage current. This was further confirmed by SEM images of the PRGO/P(VDF-TrFE-CFE) film (Fig. S4 in ESI), from which no phase separation was observed, indicating the uniform dispersion and good compatibility of the PRGO nanosheets and P(VDF-TrFE-CFE).

The Raman spectra of 4 wt% GO/P(VDF-TrFE-CFE) before and after reduction were also measured to characterize the reduction of GO, as shown in Fig. 2(c). Before reduction, the D band at 1349 cm^{-1} should be assigned to the breathing modes of the hexagonal rings, which are closely related to the edge defects.^[48,50] The G band at 1599 cm^{-1} of the composites was assigned to the stretching of sp^2 carbons, which is related to the highly ordered graphite structures.^[48,50] The intensity ratio of the D and G bands (I_D/I_G) was 0.90. After thermal reduction, the D and G bands shift to 1354 and 1597 cm^{-1} , respectively, which is caused by the reduced domain size of the sp^2 carbons. The I_D/I_G ratio of PRGO also increases to 0.94 (see Table S4 in ESI) owing to the restoration of sp^2 carbons after the removal of oxygen-containing species.^[48] The limited change in the I_D/I_G ratio also indicates a relatively low degree of reduction. The broad and low peak at about 2700 cm^{-1} was assigned to the 2D band, indicating that GO and PRGO were probably stacked as multilayers in the polymer matrices.^[50] The brown films became dark after the thermal reduction, as illustrated in the inset photographs in Fig. 2(c). The XPS spectra of the PRGO films with different reduction times are shown in Fig. 2(d) and the C 1s peak increased significantly after reduction. The C/O ratio calculated from the XPS results increase from 2.14 (0 days) to 3.70 (12 days), as shown in the figure. The representative deconvoluted results of the C 1s orbitals are shown in Figs. 2(e) and 2(f), from which an intense increase in the carbon-carbon double bond could be observed. From the F 1s XPS spectra (Fig. 2g), we can observe a notable displacement of the peak from 686.8 eV to 690.0 eV , indicating a decreased electron density on the F atom, which is probably caused by hydrogen bonding between the filler and polymer.^[51]

All data support the conclusion that GO is partially reduced and that interactions between the filler and polymer matrix exist. The restoration of sp^2 carbons results in an enhancement of the conjugation degree and conductivity of GO, which accounts for the increase in the dielectric constant. Additionally, the limited degree of reduction could significantly inhibit the long-range transport and migration of charge carriers in the composites, which is responsible for the low loss. In addition, the oxygen-containing functional groups of GO facilitate stable and homogenous dispersion through microscopic interactions, and the *in situ* thermal reduction could maintain the distribution to the maximum extent. The influ-

ence of the total dipole moment of the dielectric composite before and after reduction was also evaluated by molecular dynamic (MD) simulation, which showed a 3.4-fold increase in the total dipole moment after the reduction of GO (from 17.7 D to 60.1 D), as shown in Fig. 2(h). The partial removal of oxygen-containing functional groups and extended π -conjugation of the filler after reduction resulted in an increase in the total dipole moment, thereby enhancing the dielectric constant. This result is consistent with the experimental finding that the relative dielectric constant increases from 74 to 1415 (19.1 times). The growth rate of the simulation result is not as significant as the experimental result, probably because the chemical structures, defects, and sizes of the GO and PRGO used in the simulation are different from those used in practical applications. The time-dependent dipole moment change reflected the orientation of the polar domains in the composites.^[52] The snapshots of the composites and the corresponding chemical structures used in the simulation are shown in Fig. S5 (in ESI) and the orientation required approximately 25 ps to achieve equilibrium in both the dielectric PMCs. The orientation process involves the rotation and rearrangement of molecules and should overcome the resistance from microscopic interactions.^[53,54] The relatively long time required for the polarization process provides evidence of strong interactions between the filler and the matrix. The simulation results indicate that changes in the chemical structures and molecular interactions can affect the dipole moment in PMC, thereby modulating the dielectric properties.

The thermal reduction of GO is generally accompanied by the removal of oxygen-containing groups as gases (e.g. O_2 , CO, and CO_2), and is an exothermic reaction.^[55] The Reduction of GO is generally conducted at relatively high temperatures (typically $>350\text{ }^\circ\text{C}$).^[55,56] Considering the relatively low reduction temperature and based on the results of FTIR and Raman spectra, it could be concluded that the GO nanosheets in the P(VDF-TrFE-CFE) polymers were only partially reduced, although they had already reached equilibrium at a reduction temperature of $120\text{ }^\circ\text{C}$. The chemical reduction of GO by Vitamin C (V_C -RGO)^[57] was also conducted for comparison. The V_C -RGO composite exhibits a 2-fold higher dielectric constant but an approximate 17.5-fold higher loss than the thermally reduced GO (T-RGO) composite at 100 Hz (see Fig. S6 in ESI). V_C -RGO exhibits a higher conductance (G) than T-RGO, and chemical reduction generally introduces more defects. A similar observation was made in the PRGO/PVA composite with a reduction temperature of $160\text{ }^\circ\text{C}$, in which a higher reduction temperature may also result in more defects and an increased G. The lower permittivity of the PRGO/PVA composite compared with P(VDF-TrFE-CFE) can be mainly attributed to the relatively low dielectric constant of PVA, which may result in weaker interfacial polarization and less efficient dipole alignment in the matrix. Because a higher reduction temperature of $160\text{ }^\circ\text{C}$ was used for the PRGO/PVA composite, the reduction degree of GO was improved and led to enhanced electrical conductivity, facilitating the formation of conductive pathways, thereby resulting in a larger dielectric loss. The pronounced hygroscopicity of PVA coupled with the potential presence of residual ions may contribute to the elevated dielectric loss observed in this system. Using high-polarity

polymers, such as P(VDF-TrFE-CFE), which are prone to interact with fillers, could facilitate the achievement of high-performance dielectric PMCs. These results imply the significance of microscopic structure control and consequently filler-matrix interaction regulation in dielectric properties for consideration of the trade-off between the dielectric constant and loss.

Proximity Sensor

Proximity sensors can detect the movement of objects without any direct contact and have shown promising applications in security control, human-machine interaction, and robotics.^[58,59] Among all types of proximity sensors, capacitive proximity sensors are attractive because of their high sensitivity, low cost, and ease of fabrication. The electrode design of capacitive proximity sensors is an effective method of enhancing the fringe field to solve the crucial problems of low sensitivity and poor perception.^[60] Using a high- k dielectric layer can theoretically improve the sensitivity and reliability of capacitive proximity sensors. To verify this, models of capacitive proximity sensors composed of PRGO/P(VDF-TrFE-CFE) and polyimide (PI) were developed and studied. In these models, the dielectric constants of PRGO/P(VDF-TrFE-CFE) and PI were 1415 and 2.8, respectively. The simulation results show that the original capacitance (C_0) of the PRGO/P(VDF-TrFE-CFE) sensor was about 56-time higher than that of the polyimide (PI) sensor, as shown in Fig. 3(a). Meanwhile, the capacitance change (ΔC) of the high- k composite based sensor is approximate 1.1–3.5 times of that of PI-based one, when the distance between the material under test (MUT) and sensor is less than 5 cm. Consequently, compared with the PI-based sensor, the relative capacitance change ($\Delta C/C_0$) of the sensor made of high- k materials was much smaller. This indicates the reliability and stability of capacitive proximity sensors fabricated using high- k materials.

Apart from the dielectric properties of the functional layer, the number, arrangement, and geometry of the electrodes are key issues in sensor design. Most studies have focused on the effect of electrode design on the capacitance (C) and/or capacitance change (ΔC) of proximity sensors that vary with the approach of the MUT. In practice, $\Delta C/C_0$ is often applied to detect the distance between the MUT and sensor. However, the variation in the ratio of electrode width to distance between electrodes has rarely been systematically studied to enhance $\Delta C/C_0$ of interdigitated sensors under MUT approaches. Analytical and numerical methods were utilized to deepen the understanding of the arrangement and geometry influencing the performance of capacitive proximity sensors, as well as to direct the design of appropriate electrode patterns (see the section of Analytical evaluation in ESI). Based on the simulation results (see Figs. S7–S13 in ESI) and considering the cost, sensor size, environmental benefits, and manufacturing process, the sensors in this research were designed in a unit structure with d/w of 5 (1 cm/0.2 cm) to obtain a large relative capacitance change. A polymer composite was fabricated on the surface of the PI film coated with the electrodes. A schematic illustration of the sensor and the equivalent circuit is shown in Figs. 3(b) and 3(c). A photograph of the sensor is shown in Fig. S14 (in ESI). When an object approaches the sensor, the sensor operates in the shunt mode.^[59] Generally, a low potential exists between the object and the ground, which results in the capacitance between them (C_{OG}). Capacitances were also present between the four electrodes (C_{12} , C_{23} and C_{34}), as well as between the object and each electrode (C_{10} , C_{20} , C_{30} and C_{40}). An object approaching a sensor attenuates the electric fields between the electrodes owing to the presence of C_{10} , C_{20} , C_{30} and C_{40} . This resulted in a decrease in C_{12} , C_{23} and C_{34} . Figs. 3(d) and 3(e) illustrate the cross-sectional and top-view electrical potential

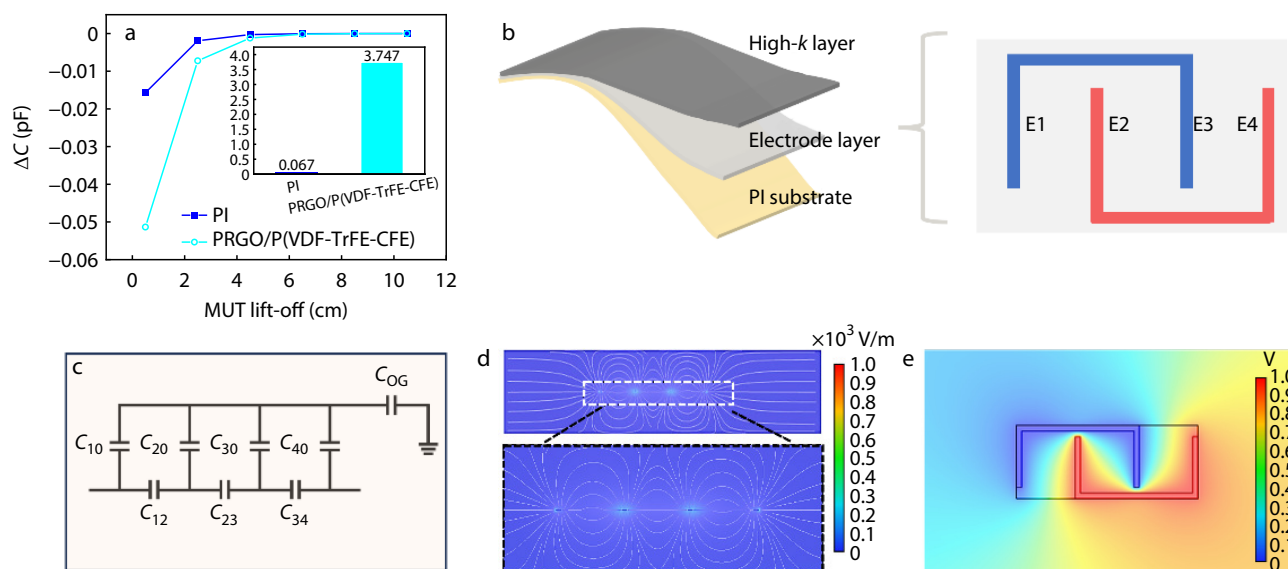


Fig. 3 Simulation results. (a) The capacitance (inserted) and capacitance change of sensors composed of PI and PRGO/P(VDF-TrFE-CFE) with dielectric constant of 2.8 and 1415, respectively; (b) Sensor and electrode structure; (c) The equivalent circuit of an object approaching the sensor with optimized electrode design. C_{OG} is the capacitance between the object and ground; C_{12} , C_{23} and C_{34} are the capacitance between electrodes; C_{10} , C_{20} , C_{30} and C_{40} represent the capacitance between the object and each electrode; (d) Cross-sectional electric field and (e) the electrical potential of the optimized sensor.

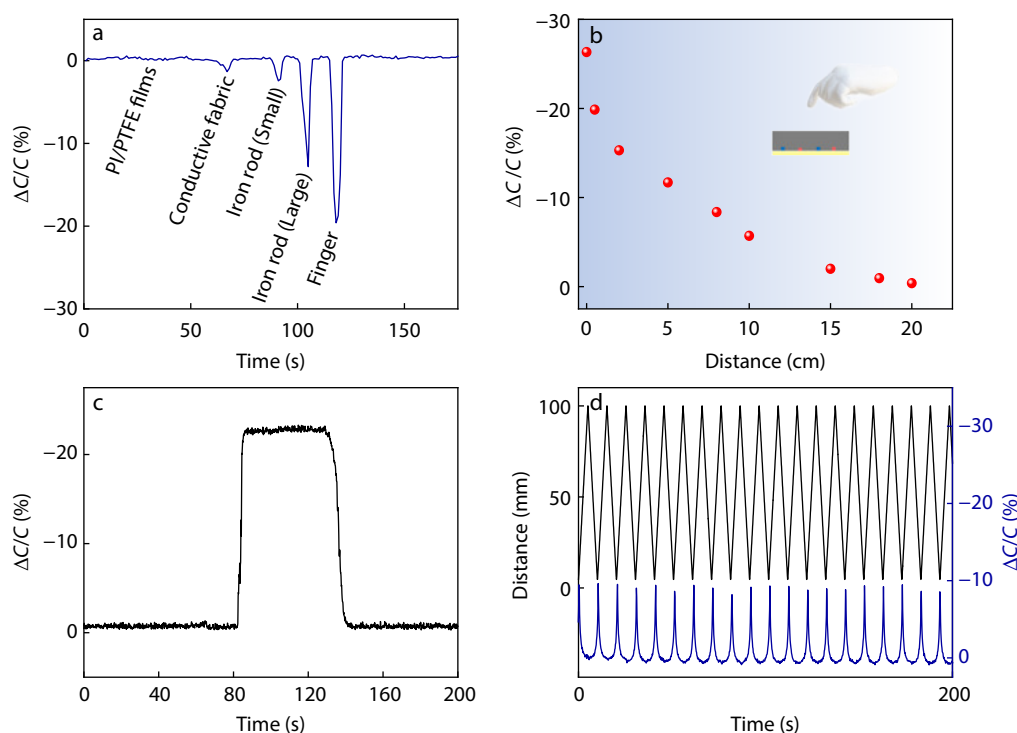


Fig. 4 Capacitive proximity sensor performance. (a) Sensor responses to different objects, including insulating polymer films (PI and PTFE, 20 cm $\times$ 2 cm $\times$ 50 μ m), conductive fabric (20 cm $\times$ 2 cm $\times$ 150 μ m), iron rod small ($D=2$ cm, $L=10$ cm) iron rod large ($D=3$ cm, $L=50$ cm) and human finger; (b) Capacitance change with the proximity of human finger; (c) Static response of the sensor to human finger (5 mm and >20 cm); (d) Cycle stability of 20 cycles.

distributions of the optimized sensor.

The sensor could respond to the proximity of conductive materials, such as metals and human fingers, but was not affected by insulating polymers (Fig. 4a). In addition to the materials, the capacitance change of the sensor is also related to the sample size (Figs. S10 and S11 in ESI). The maximum detection distance for the finger approach was 20 cm, and the sensitivity increased rapidly when the distance was less than 5 cm. The capacitance exhibited a large decrease of 20 % at 5 mm and about 26% when the finger touched the sensor, as shown in Fig. 4(b). Fig. 4(c) illustrates the static response of the human finger at 5 mm for 60 s, which remained almost constant with and without the proximity of the finger. To investigate the cycle stability, the sensor was placed on an Instron machine and an iron rod was used as the MUT. The distance between the rod and the sensor varied from 0.5 cm to 10 cm. It should be noted that in this case, the capacitance change was much lower than that of the finger. At 0.5 cm, the change is approximately 10%. The time required for each cycle was 10 s, and the sensor exhibited good stability between cycles, as shown in Fig. 4(d). The sensor remained stable even after 1000 cycles owing to the non-contact mechanism (Fig. S15 in ESI). The difference between each test may arise from the relatively long sampling intervals of the capacitance measurements. A comparison with representative state-of-the-art capacitive proximity sensors in terms of electrode structure, functional materials, detection distance, sensitivity, and stability, with the human hand as the tested object, is shown in Table S6 (in ESI).

CONCLUSIONS

In summary, the PRGO/P(VDF-TrFE-CFE) PMC with tunable dielectric properties, exhibiting an optimized dielectric constant of 1415 and $\tan\delta$ of 0.380 at 100 Hz, was successfully prepared through *in situ* thermal reduction of GO within the polymer matrix. The relative dielectric constant increased monotonically with an increase in the conductivity of GO after reduction, showing a high similarity in their growth trends. The limited reduction degree and uniform dispersion of GO in the polymer matrix could effectively suppress the formation of conductive pathways, thus minimizing dielectric loss. A controlled reduction process can achieve an optimal balance between a high- k and low loss. This regulatory strategy has been successfully applied to both organic and aqueous systems. Furthermore, a planar-structured capacitive proximity sensor with excellent sensitivity and stability was achieved through the systematic optimization of the electrode configuration by numerical simulation. The sensor displayed a capacitance change of 20% when the finger was positioned at a 5 mm distance, indicating the good sensitivity. It also exhibited excellent stability over 1000 cycles owing to its non-contact feature. This study provides an easy and effective strategy to overcome the difficulty of simultaneously realizing a high dielectric constant with low dielectric loss in a controllable manner, which is of significance in the field of flexible and wearable electronics, as well as energy devices.

Conflict of Interests

The authors declare no interest conflict.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at <http://doi.org/10.1007/s10118-025-3470-z>.

Data Availability Statement

The data supporting the findings of this study are available from the corresponding author upon reasonable request.

ACKNOWLEDGMENTS

This study was financially supported by the Innovation and Technology Commission of the Hong Kong SAR Government (No. MRP/020/21), Hong Kong Polytechnic University (No. 847A), RI-Wear Seed Fund of PolyU (1-CD8J), and Start-up Fund of PolyU (1-BD49).

REFERENCES

- Li, Y.; Luo, Y.; Deng, H.; Shi, S.; Tian, S.; Wu, H.; Tang, J.; Zhang, C.; Zhang, X.; Zha, J. W.; Xiao, S. Advanced dielectric materials for triboelectric nanogenerators: principles, methods, and applications. *Adv. Mater.* **2024**, 2314380.
- Chen, J.; Pei, Z.; Chai, B.; Jiang, P.; Ma, L.; Zhu, L.; Huang, X. Engineering the dielectric constants of polymers: from molecular to mesoscopic scales. *Adv. Mater.* **2023**, 2308670.
- Dang, Z. M.; Yuan, J. K.; Zha, J. W.; Zhou, T.; Li, S. T.; Hu, G. H. Fundamentals, processes and applications of high-permittivity polymer-matrix composites. *Prog. Mater. Sci.* **2012**, 57, 660.
- Shi, Y.; Askounis, E.; Plamthottam, R.; Libby, T.; Peng, Z.; Youssef, K.; Pu, J.; Pelrine, R.; Pei, Q. A processable, high-performance dielectric elastomer and multilayering process. *Science* **2022**, 377, 228–232.
- Tan, Y. J.; Godaba, H.; Chen, G.; Tan, S. T. M.; Wan, G.; Li, G.; Lee, P. M.; Cai, Y.; Li, S.; Shepherd, R. F.; Ho, J. S. A transparent, self-healing and high-k dielectric for low-field-emission stretchable optoelectronics. *Nat. Mater.* **2020**, 19, 182–188.
- Guo, W.; Ma, Z.; Luo, Y.; Chen, Y.; Yue, Z.; Li, L. Structure, defects, and microwave dielectric properties of Al-doped and Al/Nd co-doped Ba₄Nd_{9.33}Ti₁₈O₅₄ ceramics. *J. Adv. Ceram.* **2022**, 11, 629.
- Wei, J.; Zhu, L. Intrinsic polymer dielectrics for high energy density and low loss electric energy storage. *Prog. Polym. Sci.* **2020**, 106, 101254.
- Huang, X.; Sun, B.; Zhu, Y.; Li, S.; Jiang, P. High-k polymer nanocomposites with 1D filler for dielectric and energy storage applications. *Prog. Mater. Sci.* **2019**, 100, 187.
- Yang, W.; Lin, S.; Gong, W.; Lin, R.; Jiang, C.; Yang, X.; Hu, Y.; Wang, J.; Xiao, X.; Li, K.; Li, Y.; Zhang, Q.; Zhang, J.; Ho, S.; Liu, Y.; Hou, C.; Wang, H. Single body-coupled fiber enables chipless textile electronics. *Science* **2024**, 384, 74.
- Guan, X.; Xu, B.; Gong, J. Hierarchically architected polydopamine modified BaTiO₃@P(VDF-TrFE) nanocomposite fiber mats for flexible piezoelectric nanogenerators and self-powered sensors. *Nano Energy* **2020**, 70, 104516.
- Sharma, S.; Chhetry, A.; Sharifuzzaman, M.; Yoon, H.; Park, J. Y. Wearable capacitive pressure sensor based on MXene composite nanofibrous scaffolds for reliable human physiological signal acquisition. *ACS Appl. Mater. Interfaces* **2020**, 12, 22212.
- Rahmanudin, A.; Tate, D. J.; Marcial-Hernandez, R.; Bull, N.; Garlapati, S. K.; Zamhuri, A.; Khan, R. U.; Faraji, S.; Gollu, S. R.; Persaud, K. C.; Turner, M. L. Robust high-capacitance polymer gate dielectrics for stable low-voltage organic field-effect transistor sensors. *Adv. Electron. Mater.* **2020**, 6, 1901127.
- Zhang, J.; Hao, Y.; Liu, Y.; Wang, R.; Guo, L.; Cai, Z.; Bi, K. Space charge regulated high-k polymer nanocomposite with a novel sandwich structure. *Compos. Part B Eng.* **2020**, 203, 108461.
- Hu, H.; Zhang, F.; Luo, S.; Chang, W.; Yue, J.; Wang, C. H. Recent advances in rational design of polymer nanocomposite dielectrics for energy storage. *Nano Energy* **2020**, 74, 104844.
- Tian, M.; Yan, H.; Sun, H.; Zhang, L.; Ning, N. Largely improved electromechanical properties of thermoplastic dielectric elastomers by grafting carboxyl onto SBS through thiol-ene click chemistry. *RSC Adv.* **2016**, 6, 96190.
- Sun, H.; Jiang, C.; Ning, N.; Zhang, L.; Tian, M.; Yuan, S. Homogeneous dielectric elastomers with dramatically improved actuated strain by grafting dipoles onto SBS using thiol-ene click chemistry. *Polym. Chem.* **2016**, 7, 4072.
- Xia, W.; Lu, J.; Tan, S.; Liu, J.; Zhang, Z. In plastics design library, dielectric polymer materials for high-density energy storage, William Andrew Publishing, **2018**, p. 103–163.
- Arbatti, M.; Shan, X.; Cheng, Z. Y. Ceramic-polymer composites with high dielectric constant. *Adv. Mater.* **2007**, 19, 1369.
- Wang, G.; Huang, X.; Jiang, P. Core@double-shell structured BaTiO₃-polymer nanocomposites with high dielectric constant and low dielectric loss for energy storage application. *ACS Appl. Mater. Interfaces* **2015**, 7, 18017.
- Li, Q.; Chen, L.; Gadinski, M. R.; Zhang, S.; Zhang, G.; Li, H.; Haque, A.; Chen, L. Q.; Jackson, T.; Wang, Q.; Flexible high-temperature dielectric materials from polymer nanocomposites. *Nat. Mater.* **2015**, 523, 576.
- Zhou, W.; Cao, G.; Yuan, M.; Zhong, S.; Wang, Y.; Liu, X.; Cao, D.; Peng, W.; Liu, J.; Wang, G.; Dang, Z.; Li, B. Core-shell engineering of conductive fillers toward enhanced dielectric properties: a universal polarization mechanism in polymer conductor composites. *Adv. Mater.* **2023**, 35, 2207829.
- Dang, Z. M.; Zheng, M. S.; Zha, J. W. 1D/2D carbon nanomaterial-polymer dielectric composites with high permittivity for power energy storage applications. *Small* **2016**, 12, 1688.
- Sun, H.; Zhang, H.; Liu, S.; Ning, N.; Zhang, L.; Tian, M.; Wang, Y. Interfacial polarization and dielectric properties of aligned carbon nanotubes/polymer composites: the role of molecular polarity. *Compos. Sci. Technol.* **2018**, 154, 145.
- Zeng, Z.; Jin, H.; Chen, M.; Li, W.; Zhou, L.; Zhang, Z. Lightweight and anisotropic porous MWCNT/WPU composites for ultrahigh performance electromagnetic interference shielding. *Adv. Funct. Mater.* **2016**, 26, 303.
- Wang, J.; Wang, X.; Xu, C.; Zhang, M.; Shang, X. Preparation of graphene/poly(vinyl alcohol) nanocomposites with enhanced mechanical properties and water resistance. *Polym. Int.* **2011**, 60, 816.
- Ren, J.; Li, Q.; Yan, L.; Jia, L.; Huang, X.; Zhao, L.; Ran, Q.; Fu, M. Enhanced thermal conductivity of epoxy composites by introducing graphene@boron nitride nanosheets hybrid nanoparticles. *Mater. Des.* **2020**, 191, 108663.
- Guan, H.; Yang, R.; Li, W.; Tao, Y.; Chen, C.; Tai, H.; Su, Y.; Wang, Y.; Jiang, Y.; Li, W. Self-powered multifunctional flexible sensor for wearable biomonitoring. *Sensors Actuators B Chem.* **2023**, 377, 132996.
- Zhou, W.; Wang, Z.; Dong, L.; Sui, X.; Chen, Q. Dielectric properties and thermal conductivity of PVDF reinforced with three types of Zn particles. *Compos. Part A Appl. Sci. Manuf.* **2015**, 79, 183.
- Zhang, Y. C.; Li, W. D.; Zhao, X.; Meng, F. B.; Sun, P.; Wang, C.; Guo, G. Z.; Li, W. R.; Zhang, G. J. Epoxy-based high-k composite vitrimer: With low dielectric loss, high breakdown strength and surface electrical damage repairability. *Chem. Eng. J.* **2023**, 473,

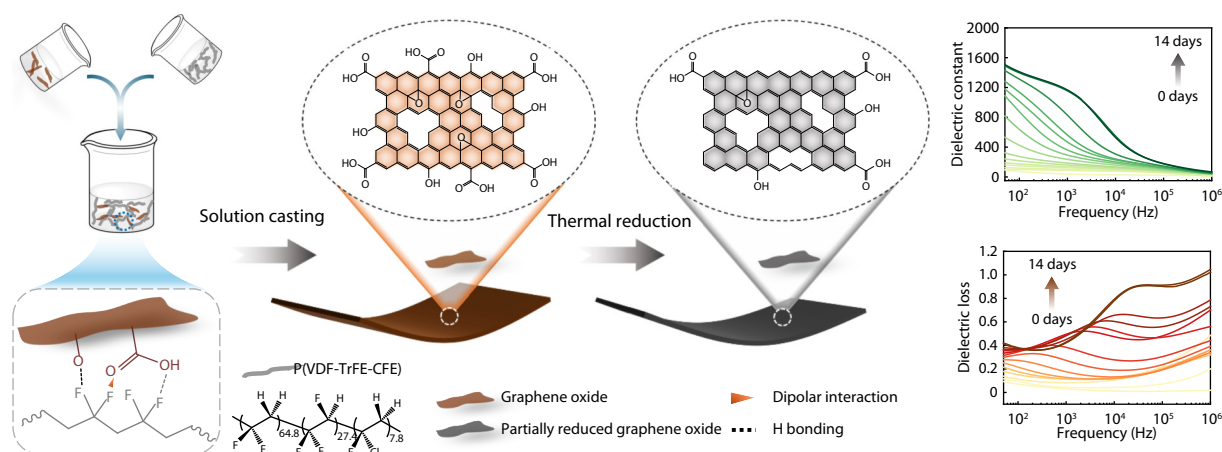
Graphical Abstract

Tailoring Dielectric Properties of Polymer Matrix Composites for High-performance Flexible Sensors

Jun Li, Kit-Ming Ma, Li-Sha Zhang, and Xiao-Ming Tao

Wuyi University; The Hong Kong Polytechnic University

A flexible dielectric ($\epsilon_r=1415$, $\tan\delta=0.380$, 100 Hz) is achieved through *in situ* controllable reduction of graphene oxide (GO) in a poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene) (P(VDF-TrFE-CFE)) matrix, arising from optimized electron conjugation, controlled charge migration, and uniform filler dispersion *via* micro-interactions.



Chinese J. Polym. Sci. 2026, 44, 116–126

<https://doi.org/10.1007/s10118-025-3470-z>

145199.

- 30 Wu, X.; Ivry, Y.; Zheng, J.; Zhang, P.; Zheng, Z.; Tan, D. Q. Dielectric loss reduction and validation of P(VDF-HFP) films via sandwiching film fabrication for capacitive energy storage. *Mater. Today Energy* **2023**, 31, 101209.
- 31 Wang, Z.; Fan, J.; Li, Y.; Wang, X.; Chen, H. High- k composites with sandwich structure by introducing a negative permittivity layer with Ni as conductive filler. *Ceram. Int.* **2019**, 45, 20128.
- 32 Shi, M.; Yang, W.; Zhang, Y.; Tan, J.; Cheng, L.; Jiao, Z.; Zhen, X. Mechanical and dielectric properties and crystalline behavior of multilayer graphite-filled polyethylene composites. *J. Appl. Polym. Sci.* **2019**, 136, 48131.
- 33 Jang, M.; Jung, D.; Kim, J.; Cho, J.; Yim, S.; Song, W.; Myung, S.; Lee, S. S.; An, K. S. Highly boosted homogeneity of polymer matrix composites filled with MXene-derived 2D titanium oxide nanosheet for high- k gate dielectrics. *J. Alloys Compd.* **2023**, 947, 169563.
- 34 Wang, Z.; Han, N. M.; Wu, Y.; Liu, X.; Shen, X.; Zheng, Q.; Kim, J. K. Ultrahigh dielectric constant and low loss of highly-aligned graphene aerogel/poly(vinyl alcohol) composites with insulating barriers. *Carbon* **2017**, 123, 385.
- 35 Luo, S.; Shen, Y.; Yu, S.; Wan, Y.; Liao, W. H.; Sun, R.; Wong, C. P. Construction of a 3D-BaTiO₃ network leading to significantly enhanced dielectric permittivity and energy storage density of polymer composites. *Energy Environ. Sci.* **2017**, 10, 137.
- 36 Li, W.; Song, Z.; Qian, J.; Tan, Z.; Chu, H.; Wu, X.; Nie, W.; Ran, X. Enhancing conjugation degree and interfacial interactions to enhance dielectric properties of noncovalent functionalized graphene/poly(vinylidene fluoride) composites. *Carbon* **2019**, 141, 728.
- 37 Meng, X.; Zhou, W.; Chen, X.; Kong, F.; Zhao, J.; Li, W.; Zhang, Y.; Wang, F.; Yuan, M. Synergistic regulation of intra-particle and inter-particle polarizations in BaTiO₃@Al₂O₃/PVDF nanocomposites towards boosted overall dielectric properties. *Mater. Today Chem.* **2025**, 43, 102492.
- 38 Zhang, M.; Zhou, W.; Zhao, S.; Zhao, J.; Li, C.; Fan, J.; Jia, Y. Decorating insulating silica shell on silicon particles enables elevated dielectric properties in silicon/poly(vinylidene fluoride) composites. *Inorg. Chem. Commun.* **2025**, 179, 114726.
- 39 Hu, H.; Liu, Y.; Chen, L.; Zhang, F. Simultaneous enhancement of mechanical robustness and self-sensing performance in fly ash-based geopolymers nanocomposites. *Compos. Part A Appl. Sci. Manuf.* **2025**, 194, 108894.
- 40 Tang, H.; Ehlert, G. J.; Lin, Y.; Sodano, H. A. Highly efficient synthesis of graphene nanocomposites. *Nano Lett.* **2012**, 12, 84.
- 41 Zhang, X. J.; Wang, G. S.; Cao, W. Q.; Wei, Y. Z.; Cao, M. S.; Guo, L. Fabrication of multi-functional PVDF/RGO composites via a simple thermal reduction process and their enhanced electromagnetic wave absorption and dielectric properties. *RSC Adv.* **2014**, 4, 19594.
- 42 Liu, S.; Tian, M.; Yan, B.; Yao, Y.; Zhang, L.; Nishi, T.; Ning, N. High performance dielectric elastomers by partially reduced graphene oxide and disruption of hydrogen bonding of polyurethanes. *Polymer* **2015**, 56, 375.
- 43 Yousefi, N.; Sun, X.; Lin, X.; Shen, X.; Jia, J.; Zhang, B.; Tang, B.; Chan, M.; Kim, J. K. Highly aligned graphene/polymer nanocomposites with excellent dielectric properties for high-performance electromagnetic interference shielding. *Adv. Mater.*

<https://doi.org/10.1007/s10118-025-3470-z>

- 2014, 26, 5480.
- 44 Zaki, H. M. AC conductivity and frequency dependence of the dielectric properties for copper doped magnetite. *Phys. B Condens. Matter* **2005**, 363, 232.
 - 45 Heikes, R. R.; Johnston, W. D. Mechanism of conduction in Li-substituted transition metal oxides. *J. Chem. Phys.* **1957**, 26, 582.
 - 46 Luo, H.; Zhou, X.; Ellingford, C.; Zhang, Y.; Chen, S.; Zhou, K.; Zhang, D.; Bowen, C. R.; Wan, C. Interface design for high energy density polymer nanocomposites. *Chem. Soc. Rev.* **2019**, 48, 4424.
 - 47 Tashiro, K.; Kobayashi, M.; Tadokoro, H. Vibrational spectra and disorder-order transition of poly(vinylidene fluoride) form III. *Macromolecules* **1981**, 14, 1757.
 - 48 Hidayah, N. M. S.; Liu, W.; Lai, C.; Noriman, N. Z.; Khe, C. S.; Hashim, U.; Lee, H. C. Comparison on graphite, graphene oxide and reduced graphene oxide: synthesis and characterization. *AIP Conf. Proc.* **2017**, 1892, 150002.
 - 49 Ning, N.; Li, S.; Sun, H.; Wang, Y.; Liu, S.; Yao, Y.; Yan, B.; Zhang, L.; Tian, M. Largely improved electromechanical properties of thermoplastic polyurethane dielectric elastomers by the synergistic effect of polyethylene glycol and partially reduced graphene oxide. *Compos. Sci. Technol.* **2017**, 142, 311.
 - 50 Ferrari, A. C.; Basko, D. M. Raman spectroscopy as a versatile tool for studying the properties of graphene. *Nat. Nanotechnol.* **2013**, 8, 235.
 - 51 Tao, M.; Liu, F.; Xue, L. Hydrophilic poly(vinylidene fluoride) (PVDF) membrane by in situ polymerisation of 2-hydroxyethyl methacrylate (HEMA) and micro-phase separation. *J. Mater. Chem.* **2012**, 22, 9131.
 - 52 Cai, F.; Luo, Y.; Yang, W.; Ye, X.; Zhang, H.; Zhu, J.; Wu, S. Study on the thermal and dielectric properties of covalently modified GO/XNBR composites. *Mater. Des.* **2021**, 198, 109335.
 - 53 Yang, H.; Cai, F.; Luo, Y.; Ye, X.; Zhang, C.; Wu, S. The interphase and thermal conductivity of graphene oxide/butadiene-styrene-vinyl pyridine rubber composites: a combined molecular simulation and experimental study. *Compos. Sci. Technol.* **2020**, 188, 107971.
 - 54 Harada, J.; Ohtani, M.; Takahashi, Y.; Inabe, T. Molecular motion, dielectric response, and phase transition of charge-transfer crystals: acquired dynamic and dielectric properties of polar molecules in crystals. *J. Am. Chem. Soc.* **2015**, 137, 4477.
 - 55 Larciprete, R.; Fabris, S.; Sun, T.; Lacovig, P.; Baraldi, A.; Lizzit, S. Dual path mechanism in the thermal reduction of graphene oxide. *J. Am. Chem. Soc.* **2011**, 133, 17315.
 - 56 Acik, M.; Lee, G.; Mattevi, C.; Chhowalla, M.; Cho, K.; Chabal, Y. J. Unusual infrared-absorption mechanism in thermally reduced graphene oxide. *Nat. Mater.* **2010**, 9, 840.
 - 57 Bo, Z.; Shuai, X.; Mao, S.; Yang, H.; Qian, J.; Chen, J.; Yan, J.; Cen, K. Green preparation of reduced graphene oxide for sensing and energy storage applications. *Sci. Rep.* **2015**, 4, 4684.
 - 58 Fattori, M.; Cardarelli, S.; Fijn, J.; Harpe, P.; Charbonneau, M.; Locatelli, D.; Lombard, S.; Laugier, C.; Tournon, L.; Jacob, S.; Romanjek, K.; Coppard, R.; Gold, H.; Adler, M.; Zirkl, M.; Groten, J.; Tschepp, A.; Lamprecht, B.; Postl, M.; Stadlober, B.; Socratous, J.; Cantatore, E. A printed proximity-sensing surface based on organic pyroelectric sensors and organic thin-film transistor electronics. *Nat. Electron.* **2022**, 5, 289.
 - 59 Arita, H. A fast optical proximity sensor skin that contains an analog computing circuit and can cover an entire link. *Adv. Robot.* **2023**, 37, 1083.
 - 60 Hu, X.; Yang, W. Planar capacitive sensors-designs and applications. *Sens. Rev.* **2010**, 30, 24.